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Structural, magnetic and magneto-resistive properties of sputter deposited Ni-Mn-Ga ferromagnetic shape memory thin films

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ABSTRACT

Ni-Mn-Ga ferromagnetic shape memory alloy thin films were deposited onto well cleaned substrates of Si (100) and glass at two different sputtering powers of 25W and 45 W in argon atmosphere at 0.015 mbar. The structural, magnetic and magneto-resistive properties of the deposited films were systematically investigated. The sputtering target $Ni_{50}Mn_{30}Ga_{20}$ (at.%) used in the present work has been prepared by vacuum induction melting technique. The prepared thin films were post-annealed at a temperature of 600°C for 1 h in vacuum. The composition and structure of the films were studied by energy dispersive x-ray analysis and x-ray diffraction techniques. The magnetic properties were studied using vibrating sample magnetometer and a.c. magnetic susceptometer. Magneto-resistive properties were measured using four-probe set-up attached with vibrating sample magnetometer. Pristine thin films are quasi crystalline while the annealed films show good crystallinity and ferromagnetism at room temperature. It was observed that the annealed films deposited at 25 W show austenite phase whereas the films deposited at 45 W show mixed phases of both austenite and martensite. Maximum magnetic transition temperature of the films investigated was found to be 327 K which is in consistent with the results of a.c. magnetic susceptibility. The thermo-magnetic curves reflect only magnetic transition and no signature of structural transition observed. The magneto-resistive properties of the thin films are found to be isotropic with a maximum negative magneto-resistance value of -0.6% at an applied magnetic field of 2.0 Tesla.

Keywords: NiMnGa shape memory thin films; Magnetic and magneto-resistive properties; Magnetic susceptometer.

Abbreviations: MEMS – Microelectro Mechanical Systems; MR – Magneto Resistive; VSM - Vibration Sample Magnetometer

1. INTRODUCTION

Heusler NiMnGa is a multifunctional smart magnetic material capable of producing large magnetic field induced strain ~10% and negative magneto-

DISCOVERY
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resistance ~5% making it suitable for shape memory magnetic-actuation and magneto-resistive applications (Hakola et al., 2004; Andriy Vovk et al., 2005; Liu et al., 2008; Banik et al., 2007; Koike et al., 2007) . The material also possesses other technologically interesting properties viz. magneto-caloric properties used for magnetic cooling, which other ferromagnetic shape memory alloys cannot fulfill (Soderberg et al., 2008). Despite the unique properties and multifunctional applications of NiMnGa bulk alloy, its brittleness is a major drawback which limits its potential use in effective actuation applications as the bending stress causes fracture failure in the material. Thin films of NiMnGa have been proposed to solve this problem as they can be bent by deforming stresses without fracture (Makoto Ohtsuka et al., 2004; Makoto Ohtsuka et al., 2008). NiMnGa thin films have already been successfully implemented in first prototypes of MEMS devices such as micro valves and micro scanners. Although considerable work has been carried on bulk alloy of Ni-Mn-Ga, systematic studies on the preparation and characterization of Ni-Mn-Ga thin films (Ahn et al., 2001; Dong et al., 1999; Dong et al., 2004; Wuttig et al., 2000; Wu et al., 2002; Castano et al., 2003; Chernenko et al., 2008; Kohl et al., 2006; Kohl et al., 2004; Liu et al., 2006; Dubowik & Goscianska, 2007) are not much. In the present work, NiMnGa thin films were prepared by d.c magnetron sputtering technique at two different sputtering powers of 25 and 45 W at 0.015 m bar argon pressure and their structural, magnetic and magneto-resistive properties were systematically investigated. The influence of sputtering power and thermal treatment on the structural, magnetic and magneto-resistive properties of the thin films are reported.

2. MATERIALS AND METHODS

The mixture of 99.9% pure Ni, 99.999% pure Mn and 99.999% pure Ga of required composition was melted using an alumina crucible under argon atmosphere by vacuum induction heating technique. The melt is transferred into a steel mould of 50mm inner diameter to obtain the bulk alloy. The as-cast bulk alloy was hot isostatically pressed to overcome the problem of porosity and low density. The sputtering targets of 50mm diameter and 1mm thick were prepared by EDM wire cutting from bulk alloy. The composition of the sputtering target is $Ni_{50}Mn_{30}Ga_{20}$.

Thin films of NiMnGa were deposited onto well cleaned Si (100) and glass substrates in a dc magnetron sputtering system (12"MSPT, HIND HIVAC, Bangalore). Thin films were deposited at a constant argon pressure of 0.015 mbar and sputtering powers of 25W and 45W respectively. The films were subjected to post-deposition heat treatment at 600^{o}C for 1h in vacuum. The structural, magnetic and magneto-resistive (MR) properties of the thin films were systematically investigated. The chemical composition in the thin films has been determined using energy dispersive x-ray analysis and the structural characteristics have been studied using x-ray diffractometer. The magnetic field dependent magnetization M (H) and the temperature dependent magnetization M (T) properties of the thin films were investigated by vibration sample magnetometer (VSM) attached with a variable temperature cryostat. Magneto-resistance properties of the films were measured using four-probe set-up attached with VSM and the temperature dependent a.c. magnetic susceptibility measurements of the thin films were studied using a magnetic susceptometer.

Table 1 Magnetic properties of d.c. magnetron sputter deposited Ni-Mn-Ga thin films

Film ID	Composition (at.%)	Substrate	Magnetizatio M_S (emu/cc)		Coercivity, (H_C (Oe)		T_C* (K)
			In Plane	Out Plane	In Plane	Out Plane	
NMGC1 (25W)	$Ni_{57.4}Mn_{27.3}Ga_{15.3}$	Si (100)	52	63	43.51	70.12	314
NMGC1 (25W)	$Ni_{57.4}Mn_{27.3}Ga_{15.3}$	Glass	208	220	38.23	33.37	Nil
NMGC3 (45W)	$Ni_{59.1}Mn_{27.4}Ga_{13.5}$	Sil (100)	148	135	17.75	26.37	315.8(302)
NMGC3 (45W)	$Ni_{61.0}Mn_{27.8}Ga_{11.2}$	Glass	332	297	33.31	33.34	327(320)

Table 2 Magnetic- resistance properties of Ni-Mn-Ga thin films

Film ID	Composition (at.%)	Substrate	B & J direction	Resistivity ρ (μΩ-cm)	MR
NMGC1 (25W)	$Ni_{57.4}Mn_{27.3}Ga_{15.3}$	Si (100)	Parallel	891	-0.46
			Perpendicular	878	-0.51
NMGC1 (25W)	$Ni_{57.4}Mn_{27.3}Ga_{15.3}$	Glass	Parallel	1236	-0.34
			Perpendicular	1236	-0.38
NMGC3 (45W)	$Ni_{59.1}Mn_{27.4}Ga_{13.5}$	Sil (100)	Parallel	230	-0.30
			Perpendicular	229	-0.30
NMGC3 (45W)	$Ni_{61.0}Mn_{27.8}Ga_{11.2}$	Glass	Parallel	978	-0.60
			Perpendicular	977	-0.61

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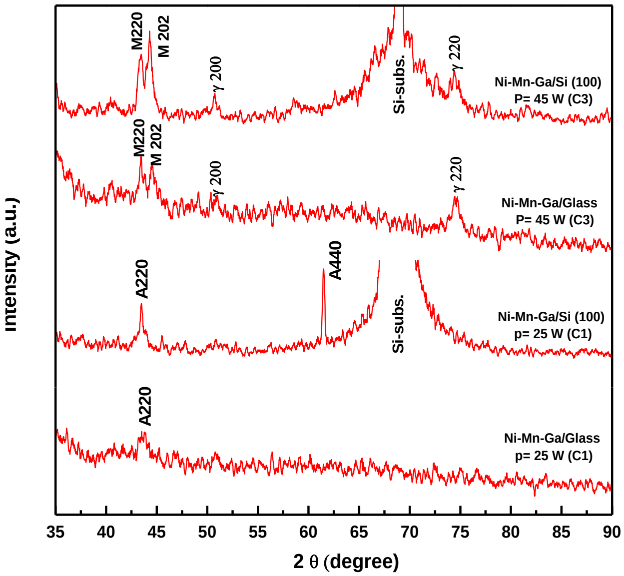

Figure 1

XRD patterns of the annealed Ni-Mn-Ga thin films

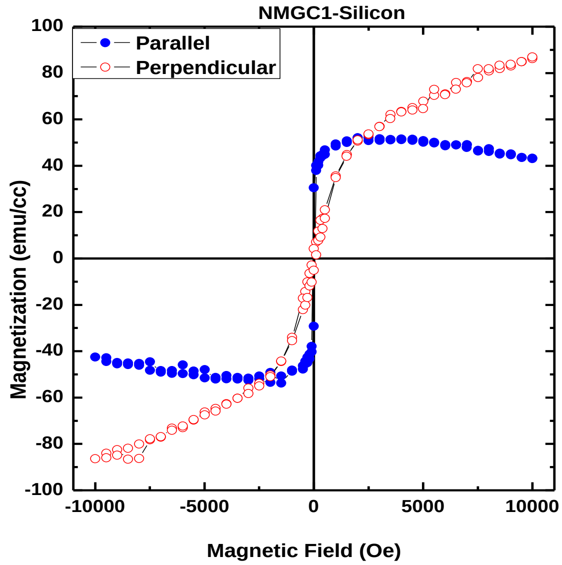

Figure 2b

M(H) hysteresis loops of annealed Ni-Mn-Ga thin films (25W).

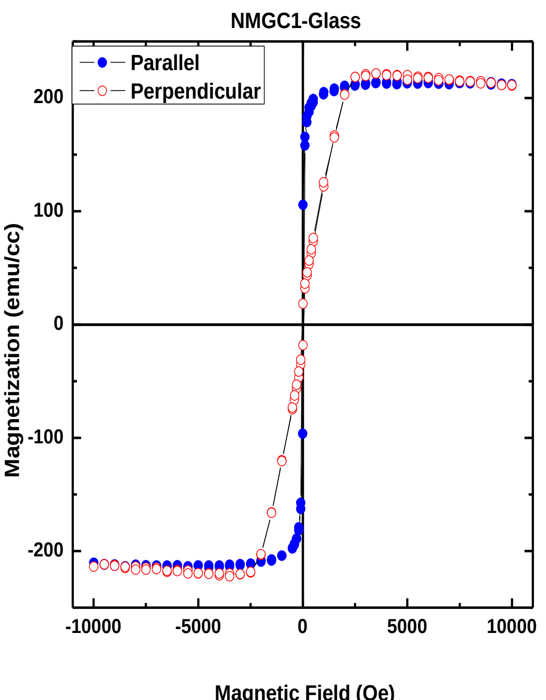

Figure 2a

M(H) hysteresis loops of annealed Ni-Mn-Ga thin films (25W)

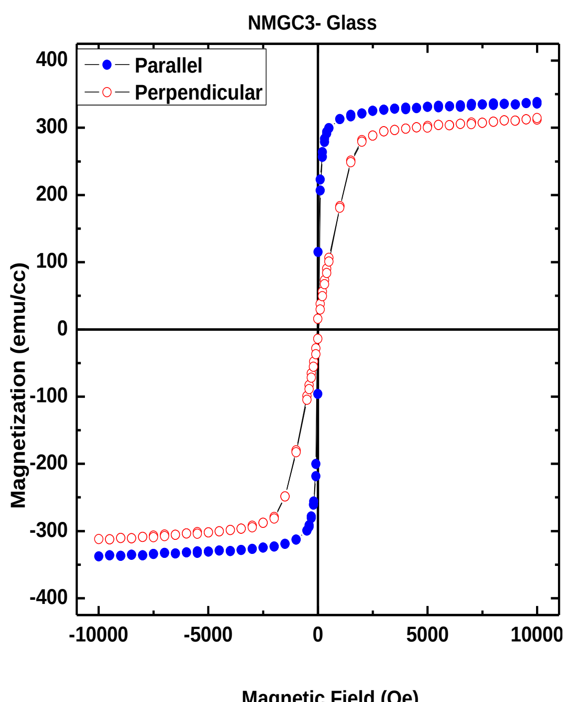

Figure 2c

M(H) hysteresis loops of annealed Ni-Mn-Ga thin films (45W)

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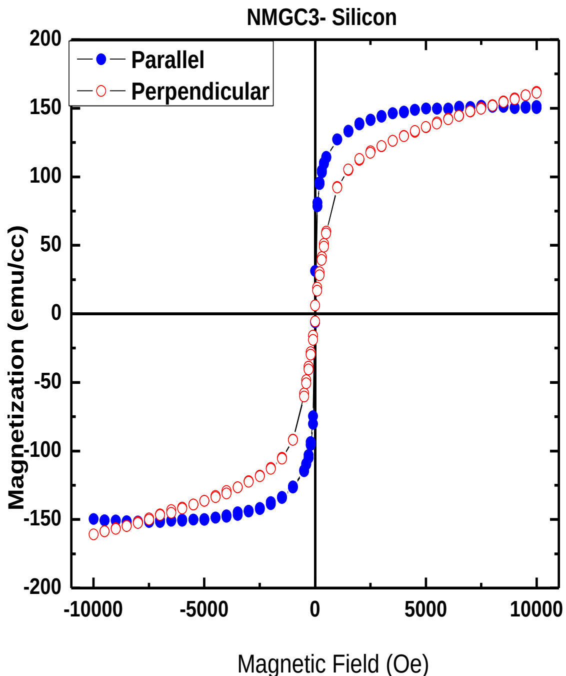

Figure 2d

M(H) hysterisis loops of annealed Ni-Mn-Ga thin films (45W)

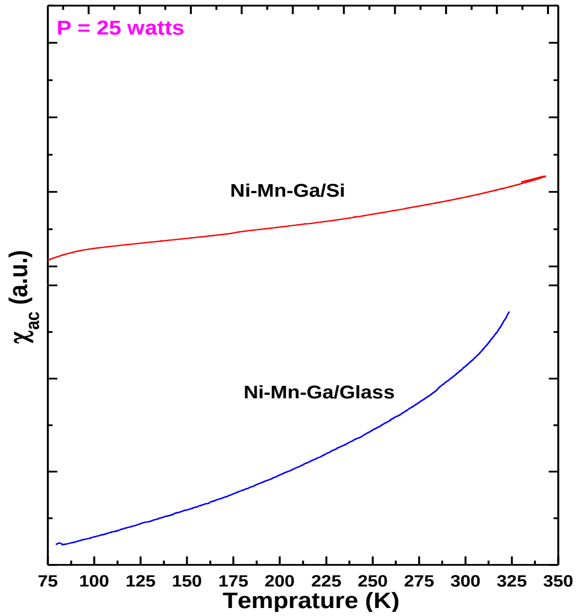

Figure 4a

A.C. magnetic susceptibility characteristics of annealed Ni-Mn-Ga thin films (25W).

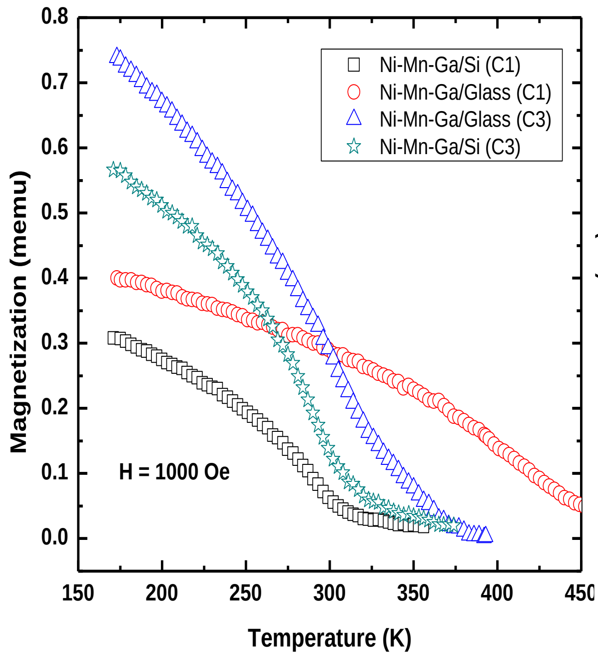

Figure 3

Thermo-magnetic curves of annealed Ni-Mn-Ga thin films

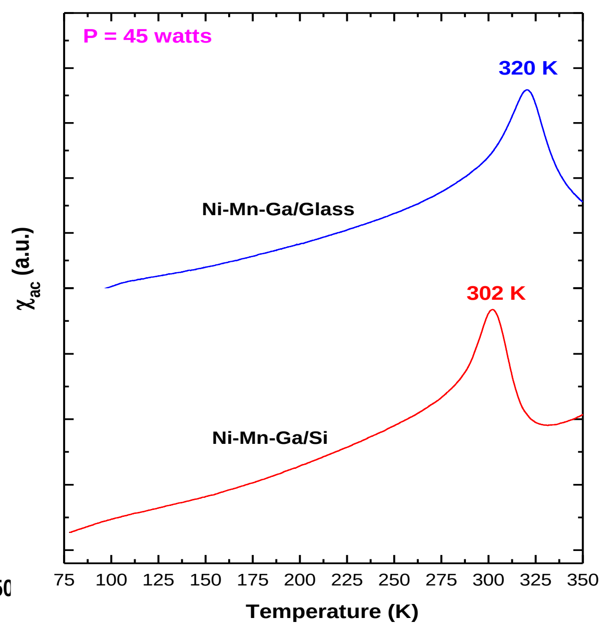

Figure 4b

A.C. magnetic susceptibility characteristics of annealed Ni-Mn-Ga thin films (45W)

3. RESULTS AND DISCUSSION

3.1. Structural analysis of NiMnGa thin films

The atomic constituents in the thin films deposited on glass and silicon substrates are given in Table 1. The structural characteristics of the pristine and post-heat treated thin films were studied by x-ray diffraction. Pristine Ni-Mn-Ga thin films are quasi-crystalline, whereas annealed films are good crystalline. The presence of well-defined peaks in diffractograms of annealed (600°C/1h) thin films shown in Fig.1 confirm the atomic ordering upon annealing. The diffraction patterns reveal the austenite phase of the annealed films deposited at 25W and the development of martensite for the films deposited at 45W. The presence of γ-phase was also observed in heat treated thin films deposited at 45W. Similar observations were also been made by Makoto Ohtsuka (2008) and Chung (2004) in NiMnGa thin films.

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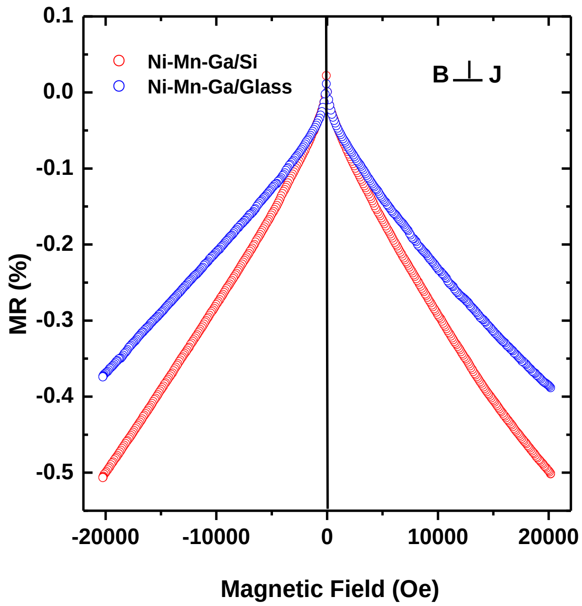

Figure 5a

Magneto-resistance curves of annealed Ni-Mn-Ga thin films (25W)

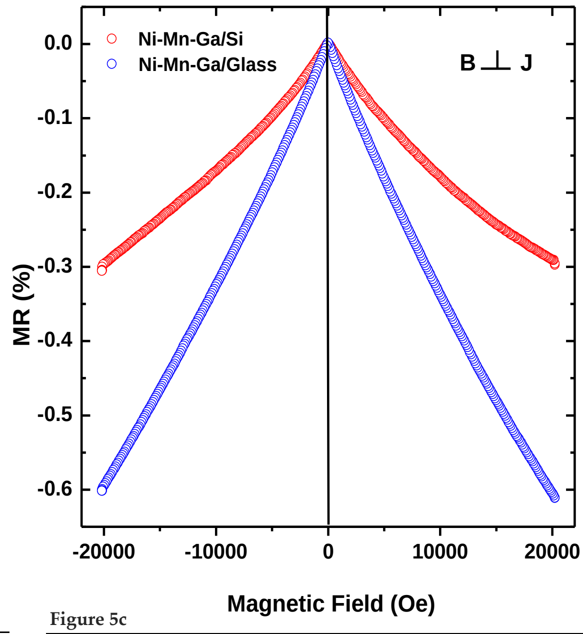

Figure 5c

Magneto-resistance curves of annealed Ni-Mn-Ga thin films (45W)

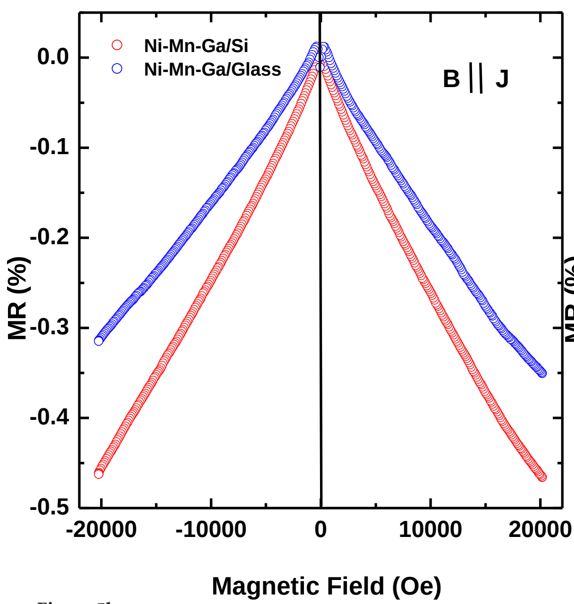

Figure 5b

Magneto-resistance curves of annealed Ni-Mn-Ga thin films (25W)

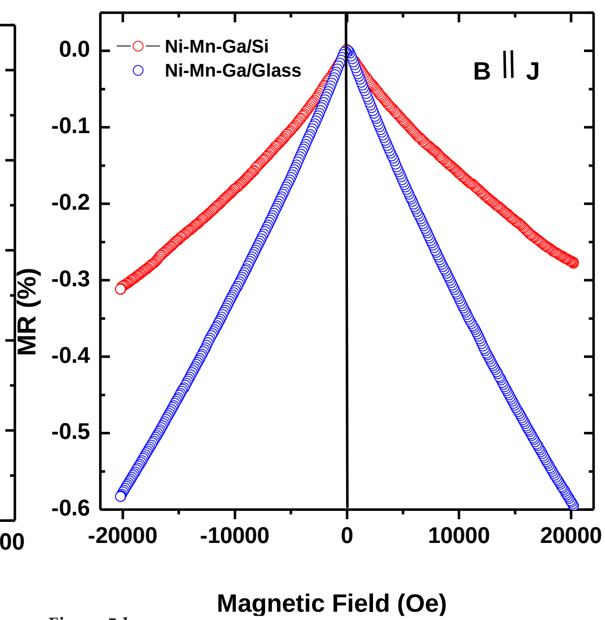

Figure 5d

Magneto-resistance curves of annealed Ni-Mn-Ga thin films (45W)

3.2. Magnetic properties of NiMnGa thin films

Figs.2a and 2b show the M (H) hysteresis loops obtained for the annealed thin films deposited at 25W on glass and si (100) substrates respectively in parallel and perpendicular directions of the film surface and magnetic field and Figs 2c and 2d show the same obtained for the films deposited at 45 W. The observation reveals that the heat treated thin films show good soft magnetic properties, characterized by narrow hysteresis loop, low coercively and high magnetic saturation. Thermo-magnetic curves obtained for the annealed thin films are shown in Fig.3, which provides the information about magnetic transition (T_c) and the values are given in Table 1. Even though information on the magnetic curie transition temperature was observed from the thermo-magnetic curves, the presence of structural transition temperature could not be identified. This may be due to small change in its magnetization during such structural transition in thin films. The curves of a.c. magnetic susceptibility obtained for the films deposited at 25W and 45 W are shown in Figs 4a and 4b respectively. The values of T_c observed from a.c susceptibility

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measurements for the heat treated thin films (45 W) are in good agreement with the values obtained from thermo-magnetic curves (Table 1).

3.3. Magneto-resistive properties of the thin films

The magneto-resistance versus applied magnetic field curves of the annealed thin films deposited at 25W and 45 W are shown in Figs 5a to 5d. The results reveal that all the samples exhibit similar magneto-resistive behaviors. The magneto resistive curves obtained for the films indicate that they show isotropic nature of magneto-resistance. The studies of magneto-resistance have been measured for both parallel and normal directions of current and magnetic field and the values are given in Table 2. A maximum negative magneto-resistance of about -0.61% has been observed for the films deposited at 45W at an applied magnetic field of about 2T. This may be due to the occurrence of ferromagnetic spin ordering of Ni-rich thin films.

4. CONCLUSION

NiMnGa bulk alloy was synthesized using induction melting technique and the sputtering targets were prepared from the bulk alloy. Thin films were prepared at two different sputtering powers of 25 and 45 W at argon pressure of 0,015 mbar using d.c. magnetron sputtering technique and their structural, magnetic, magneto-resistive and a.c. magnetic susceptibility properties were systematically investigated. Pristine thin films are found to be quasi-crystalline and paramagnetic. Well crystalline and ferromagnetic natures of the thin films were restored by post deposition heat treatment. Maximum magnetic transition temperature and maximum negative magneto-resistance values of the films investigated were found to be 327K and -0.61% respectively.

SUMMARY OF RESEARCH

1. Heusler Ni-Mn-Ga alloy is a good ferromagnetic shape memory material but brittle in nature which limits its potential applications
2. For the development of miniaturized actuators, Ni-Mn-Ga thin films are good candidates due to their high MFIS and bending nature
3. In this present work Ni-Mn-Ga films were prepared at different sputtering environments and their physical properties were investigated
4. Results reveal that the annealed films are soft ferromagnetic showing magnetic transition and no evidence of structural transition
5. Maximum magnetic transition and maximum negative magneto-resistance of the films were found to be 327 K and - 0.61% respectively.

FUTURE ISSUES

Even though signature of structural transition is absent from the thermo magnetic curves, the XRD reveals the formation of martensitic microstructure for the films deposited on si(100) which could be further addressed

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Conflicts of interests

The authors declare that there are no conflicts of interests.

Data and materials availability

All data associated with this study are present in the paper.

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